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ONR Grant No. N00014-90-J-4077

Micro-Mechanics of Electrostrictors for Sonar Transducers

Yearly Report

Period October 1, 1992–September 30, 1993

Sponsored by

Office of Naval Research (ONR)

Submitted to

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I. Executive Summary

The higher order electromechanical coupling "electrostriction" has been proposed as an alternative to the piezoelectric phenomena commonly employed for Navy-type sonar transducers. Based on the results of the parent program "Relaxor Ferroelectrics for Electrostrictive Transducers" (N00014-90-J-4077) and the three-year AASERT program on the classification of electrostrictors, it was found that the Type I - Pb(B1B2)O3 and Type II - PLZT offered superior overall performance. Specific compositions were selected based on the given criteria:

- Large E-field induced strain ≥ 0.03%
- Operating temperature range 0-30°C
- Minimal strain E-field hysteresis (<10%)

and fabricated into large ceramic rings with dimensions as follows: O.D. = 0.650 ± 0.005 in. (1.65 cm ± .013 cm), wall thickness = 0.175 ± 0.010 in. (445 cm ± 0.025 cm), and thickness = 0.281 ± 0.002 in (0.31 cm ± 0.005 cm). Ceramic rings (four each) were silver electroded and delivered to NRL in Orlando, Florida for sonar transducer evaluation.

II. Transducer Fabrication and Testing

A. Compositional Selection

According to Navy requirements, various families of electrostrictive compositions were selected for evaluation as practical Navy transducers, in order to directly compare performance with PZT piezoelectric materials.

(1) $Pb_{0.89}La_{0.11}Zr_{0.65},Ti_{0.35}O_3$ (PLZT-9/65/35), (Type II)

(2) 0.93 $Pb(Mg_{1/3}Nb_{2/3})O_3$ - 0.07 $PbTiO_3$ (PMN-PT 93/7), (Type I) and

(3) 0.85 $Pb_{0.98}La_{0.02}(Mg_{1/3}Nb_{2/3})O_3$ - 0.15 $Pb_{0.98}La_{0.02}TiO_3$ (PLMNT 2/85/15) (Hybrid I & II)

The compositional selections were based on the criteria for Navy Sonar transducers outlined in previous reports.

Dielectric data for the three compositions are presented in Table I. As seen in this Table, the PLZT-Type II electrostrictors offer the broadest region of macro-micro polar switching (T_m - T_d = 141°C),but the lowest dielectric constant (5310 @ 1 kHz). In contrast, the Type I PMN-based material exhibits much higher dielectric constants (>25,000 @ 1 kHz); however, their macro-micro polar region is much less broad. The hybrid composition (Types I & II) 2/85/15 offers an intermediate macro-micro region and dielectric permittivity. The maximum dielectric loss is similar for all three compositions (≈ 0.1 @ 1 kHz).

Table I. Dielectric data for selected electrostrictive compositions.

Composition	T_{max} (°C) (1 kHz)	T_d (°C)	T_{max} - T_d (°C)	K_{max} (1 kHz)	$\tan\delta_{max}$ (1 kHz)
PLZT (9/65/35)	77	-31	108	7,800	0.08
PMN-PT (93/7)	27.5	-9	36.5	25,000	0.095
PLMNT (2/85/15)	17.5	-49.5	67	20,500	0.10

Electric field induced transverse strain and hysteresis data for the three representative compositions are summarized in Table II. Note: Strain measurements are for the PLZT 10/65/35 composition. As seen in this Table, all three compositions exhibit induced transverse strains in excess of 0.03% (20 kV/cm) with less than 10% hysteresis within the 0 - 30°C temperature range.

Table II. Induced transverse strain data for selected electrostrictor compositions.

Composition	Temp. (°C)	Transverse Strain (%) 10 kV/cm	Transverse Strain (%) 20 kV/cm	Hysteresis (%)
PLZT (9/65/35)	24	0.50	0.072	10
	18	0.54	—	
	13	0.59	—	↓
	10	0.62	—	
	0	0.67	—	60
PMN-PT (93/7)	-8	0.032	0.042	10.2
	0	0.028	0.0395	6.4
	35	0.0215	0.0345	5.8
	-3	0.0315	0.0415	9.2
	18	0.0300	0.0415	6.5
	23	0.0270	0.0380	6.4
PLMNT (2/85/15)	5	0.0199	0.0371	6.0
	25	0.0152	0.0307	3.4
	0	0.0217	0.0392	9.1
	19	0.0136	0.0282	4.2

Note: Data given in Tables I and II are for thin disk ceramics.

B. Powder Fabrication/Synthesis

Based on the composition families (Types) given above, large quantities of ceramic powders (> 1 kg) were prepared using powder synthesis techniques developed in the parent program. The following compositions were fabricated:

(1)	$Pb_{0.9}La_{0.10}(Zr_{0.65},Ti_{0.55})O_3$	PLZT-10/65/35
	Note: 9/65/35 exhibits excessive hysteresis.	
(2)	$0.95Pb(Mg_{1/3}Nb_{2/3}$-$0.05)PbTiO_3$	PMN-PT 95/5
(3)	$0.90Pb(Mg_{1/3}Nb_{2/3}O_3$-$0.10\ PbTiO_3$ +0.5 mole% La_2O_3	PLMNT 1/10/10
(4)	$0.85Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.15\ PbTiO_3$ + 1 mole% La_2O_3	PLMNT 2/85/15

Additional formulations were synthesized, 0.93PMN-0.071PT, PLZT-10.5/65/35 and a Sr-modified PMN-PT, but were deemed insufficient in terms of overall performance. Specific issues included excessive dielectric aging and hysteresis, both believed to be associated with changes in raw materials during the course of this program.

C. Electrostrictive Ceramic Ring Fabrication

Powders prepared above were granulated and pressed into large cylinders. Subsequent to binder burnout and densification, the cylinders were machined (Piezo Kinetics, Inc.) to the desired dimensions given in the executive summary. Upon machining, the samples were electroded using a fired on silver. Special Note: The application of the silver electrode must be performed at a temperature (≤ 600°C) to minimize reactive interface which may lead to degraded dielectric performance. Unlike that found for PZT-based ceramics, the role of the electrode interface in high dielectric constant materials is of major concern.

D. Electrostrictive Ring Testing

The ceramic rings were qualified based on: (1) capacitance–temperature measurements; (2) pyroelectric behavior, and (3) polarization–E-field (P/E). The results are given in the following Figures 1-4, respectively, and summarized in Table III.

Table III. Dielectric/polarization data for selected electrostrictive compositions.

Composition	T_{max} @ 1 kHz	T_d	ΔT T_{max}-T_d	K_{max} @ 1 kHz	P @10 KV/cm
PLZT (10/65/35)	~65°C	-18°C	83°C	7000	0.085
PMN-PT (95/5)	~17°C	-25°C	42°C	27,000	0.17
PLMNT (1/90/10)	~17°C	-37°C	54°C	23,000	0.155
PLMNT (2/85/15)	~17°C	-50°C	67°C	20,000	0.155

As presented, the Type I materials had similar T_{max}s (~17°C @ 1 kHz) but with increasing La were found to have decreased K_{max}'s but expanded $\Delta T(T_{max}-T_d)$ ranges. As expected,the Type II PLZT (10/65/35) material possessed the largest ΔT (Regime II). The level of induced polarization (P_{ind}) is significantly lower than the Type I materials at 10 KV/cm; however, similar at 20 KV/cm.

In terms of hysteresis, the 95/5 samples possessed levels of < 5% @ 1 Hz, being significantly higher than that for small-thin disks samples of the same composition.

III. Scale-Up Issues

As observed for many of the compositions investigated, the level of P/E hysteresis was found to be significantly higher for the large ceramic rings in contrast to thin disks (1 cm diam., 1-2 cm thick) used in all the preliminary screening tests, Table II. In addition to hysteresis, dielectric aging was found to be prevalent. Various processing issues are known to affect dielectric aging and indirectly the P/E hysteresis due to grain-grain boundary inhomogeneity, defects, impurities, etc. The following is a list of processing parameters suggested for next generation of large sonar ring, i.e. 2" rings.

- 0.5-1% PbO deficient
- High purity raw material, i.e. Nb_2O_5
- Hot isostatic pressing
- Sr,Ca modification for grain boundary control
  — Transgranular vs. intergranular fracture

In addition to processing issues above, the following areas need to be addressed.

- Silver electrode interface

  Note: Silver electrodes used have been developed for low K PZTs.
- Uni-polar drive vs. bi-polar drive

  Note (1): Bi-polar drive gives a frequency doubling.

  Note (2): Unipolar drive may give rise to strain walk off due to the small remanent polarization tail in the temperature regime of interest.

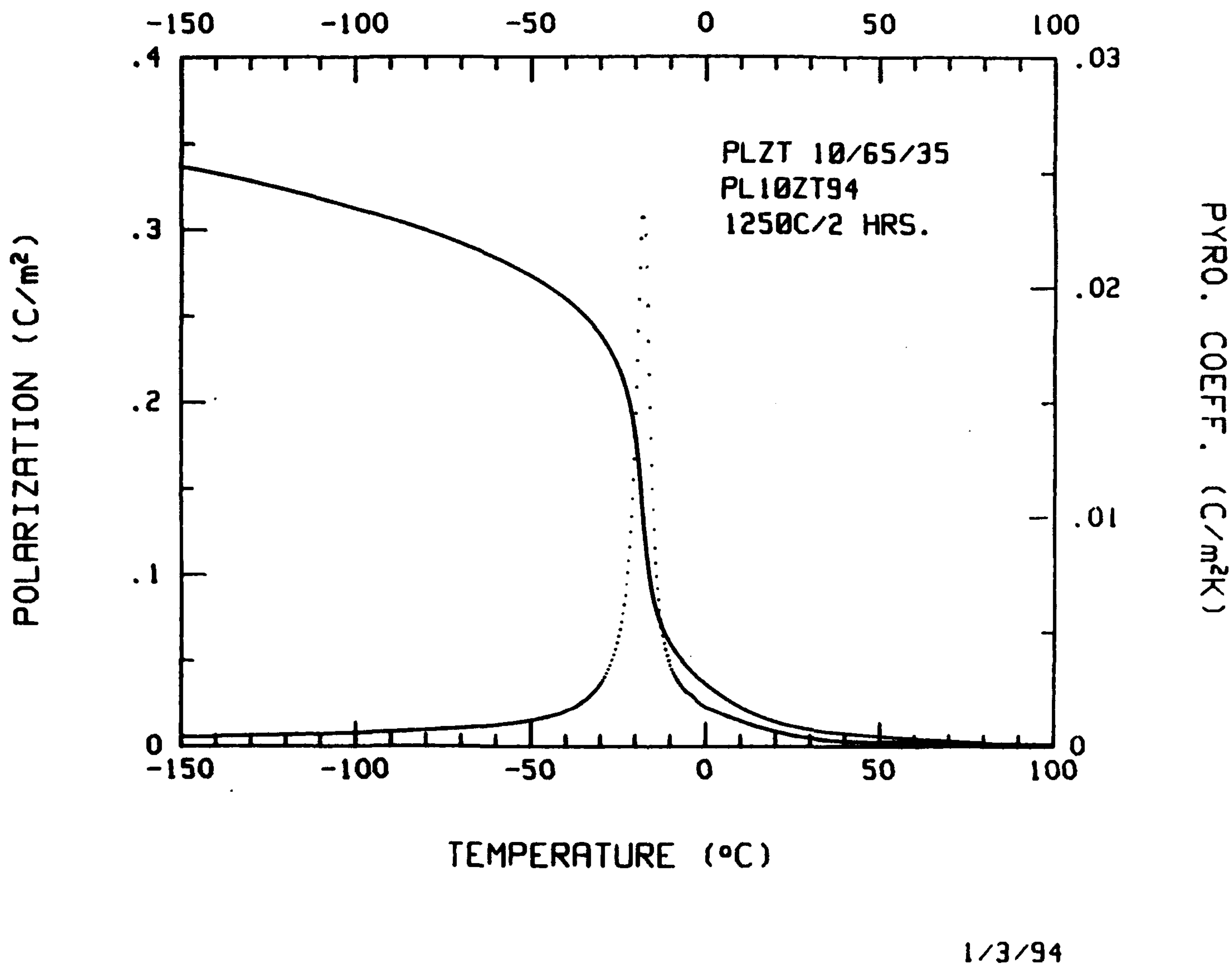


Figure 1a. Dielectric temperature behavior for PLZT 10/65/35. (Note: Frequency range 100 Hz, 1 kHz, 10 kHz, and 100 kHz.)

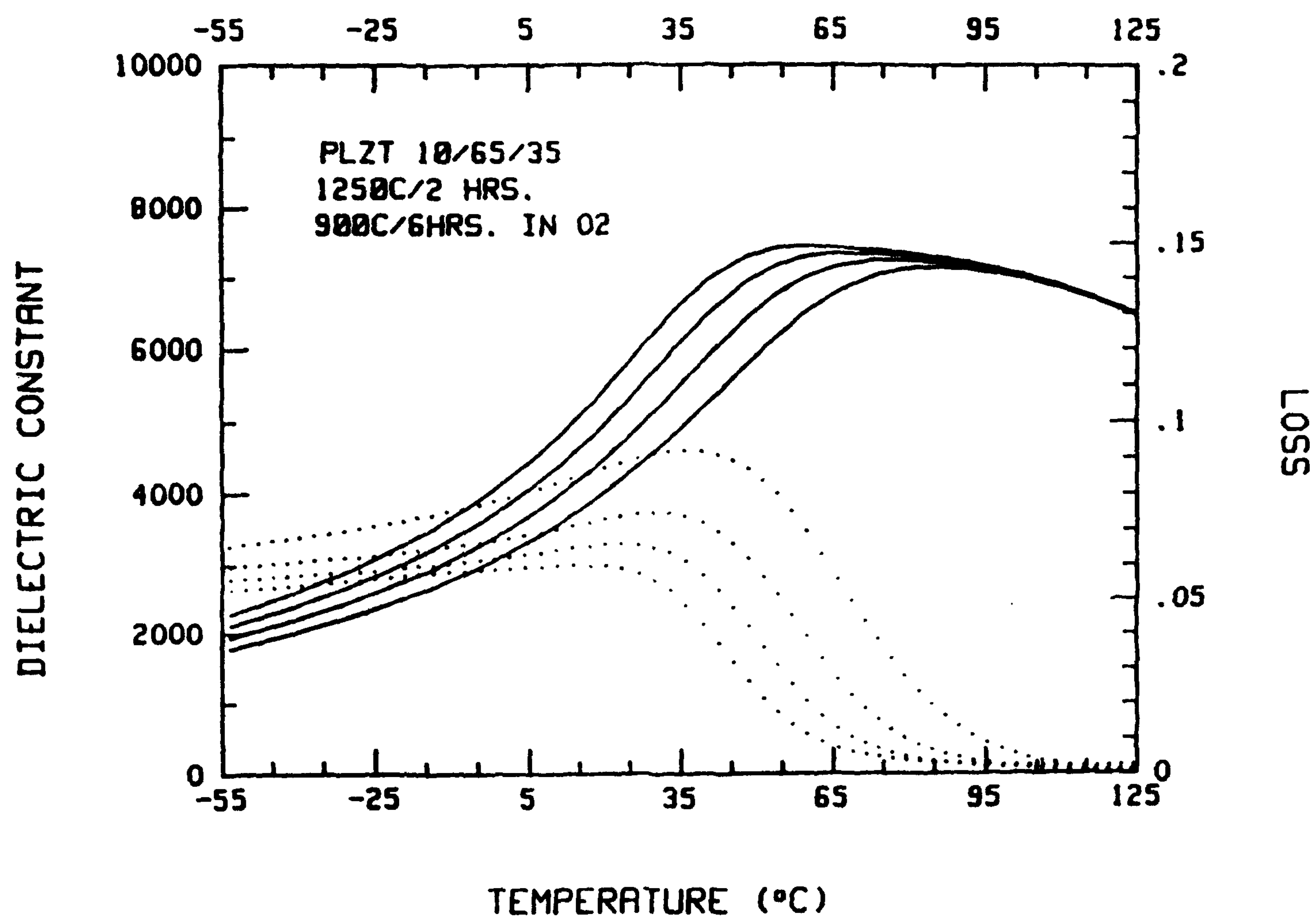
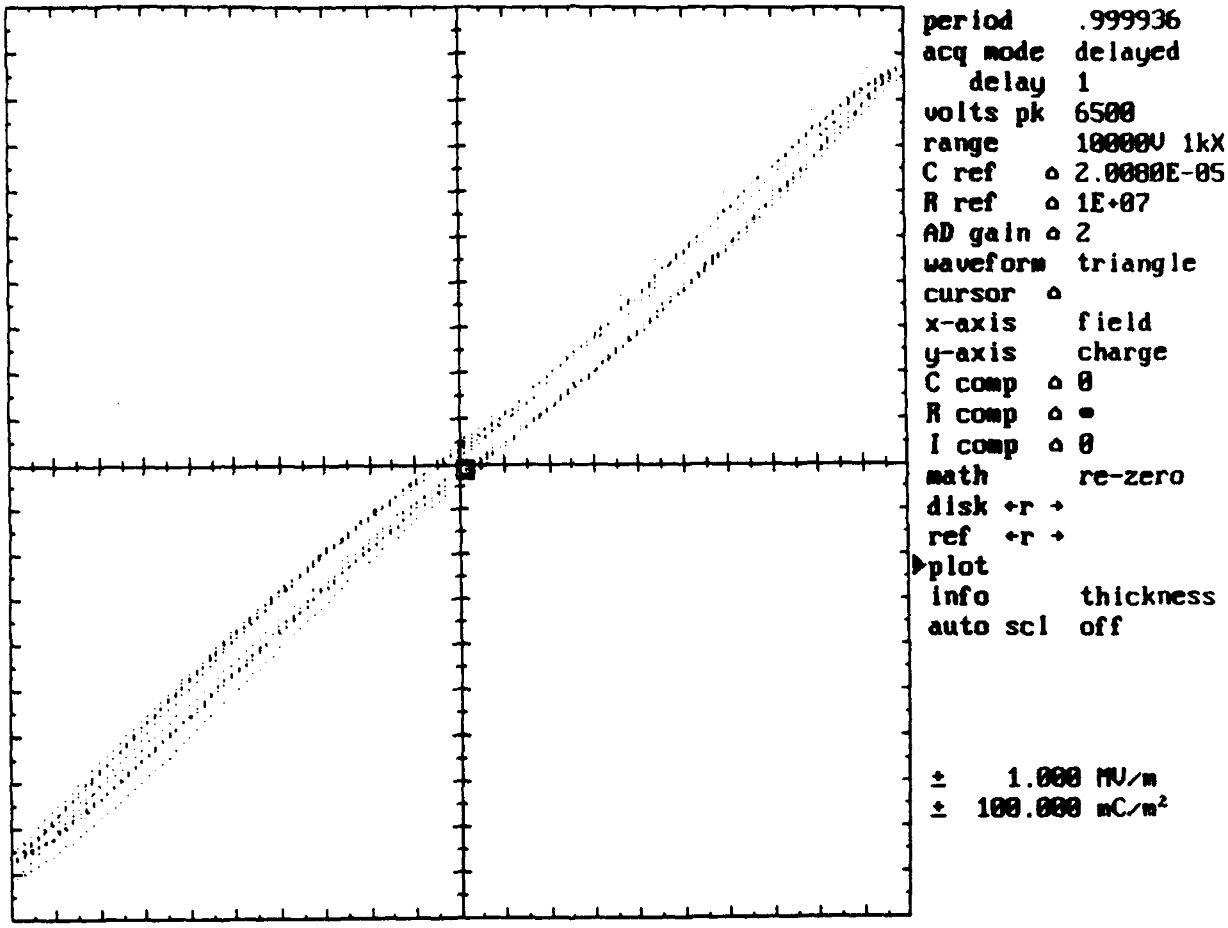


Figure 1b. Pyroelectric (dotted)/polarization (solid) temperature data for PLZT 10/65/35. (Note: T_d is designated at pyro peak temperature.



10/65/35 RT
dia =14.550 mm area= 1.663E-04 m^2 thickness=7087.000 μm

Figure 1c. **Polarization E-field behavior of PLZT 10/65/35 @ room temperature.**
Note (1): y axis - 0.1 c/m^2 full scale
x axis - 10 kV/cm full scale
Note (2): All four rings were measured as indicated by the four plots.

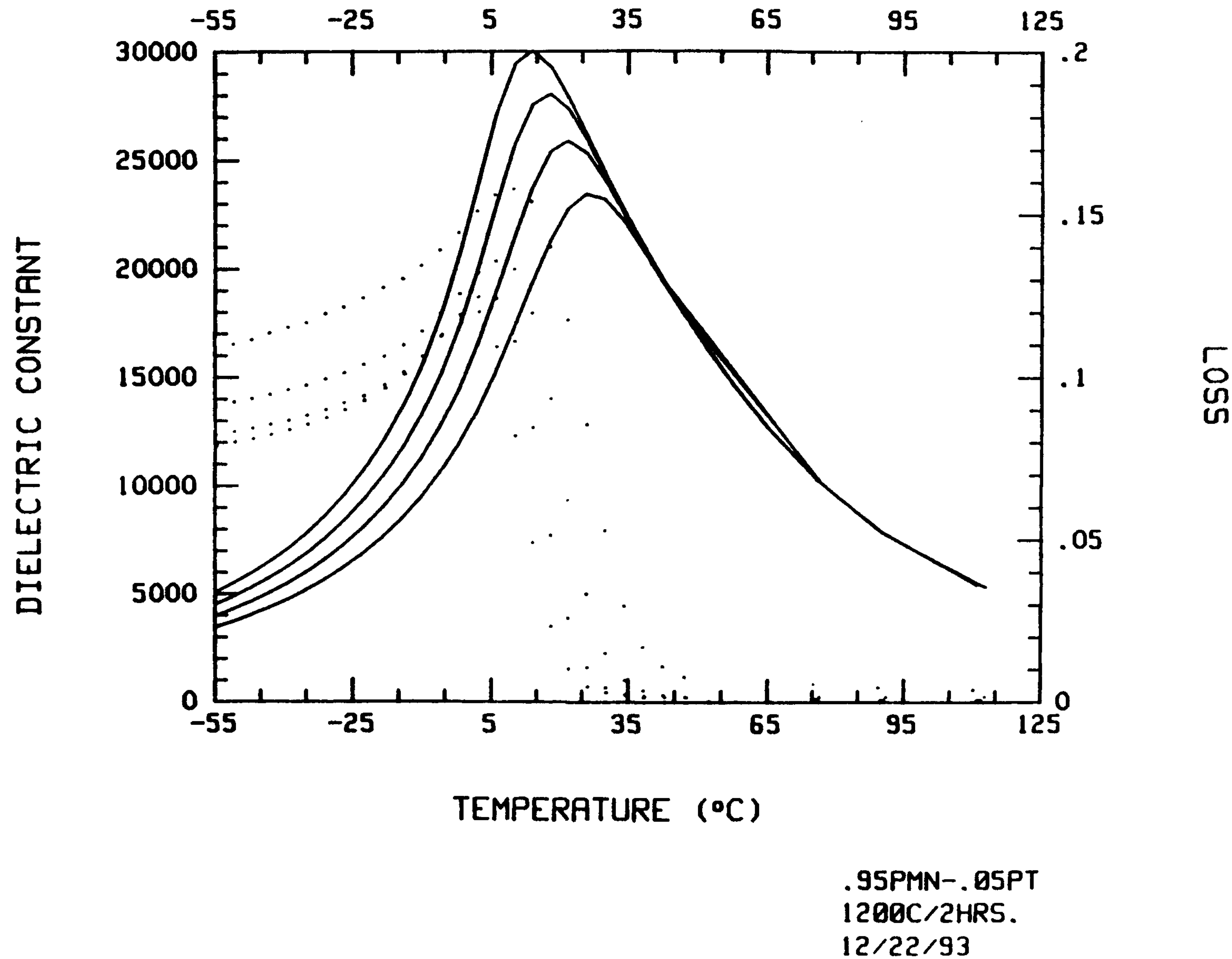


Figure 2a. Dielectric temperature behavior for 95/5 PMN:PT. Note: frequencies used 100 Hz, 1 kHz, 10 kHz, 100 kHz.

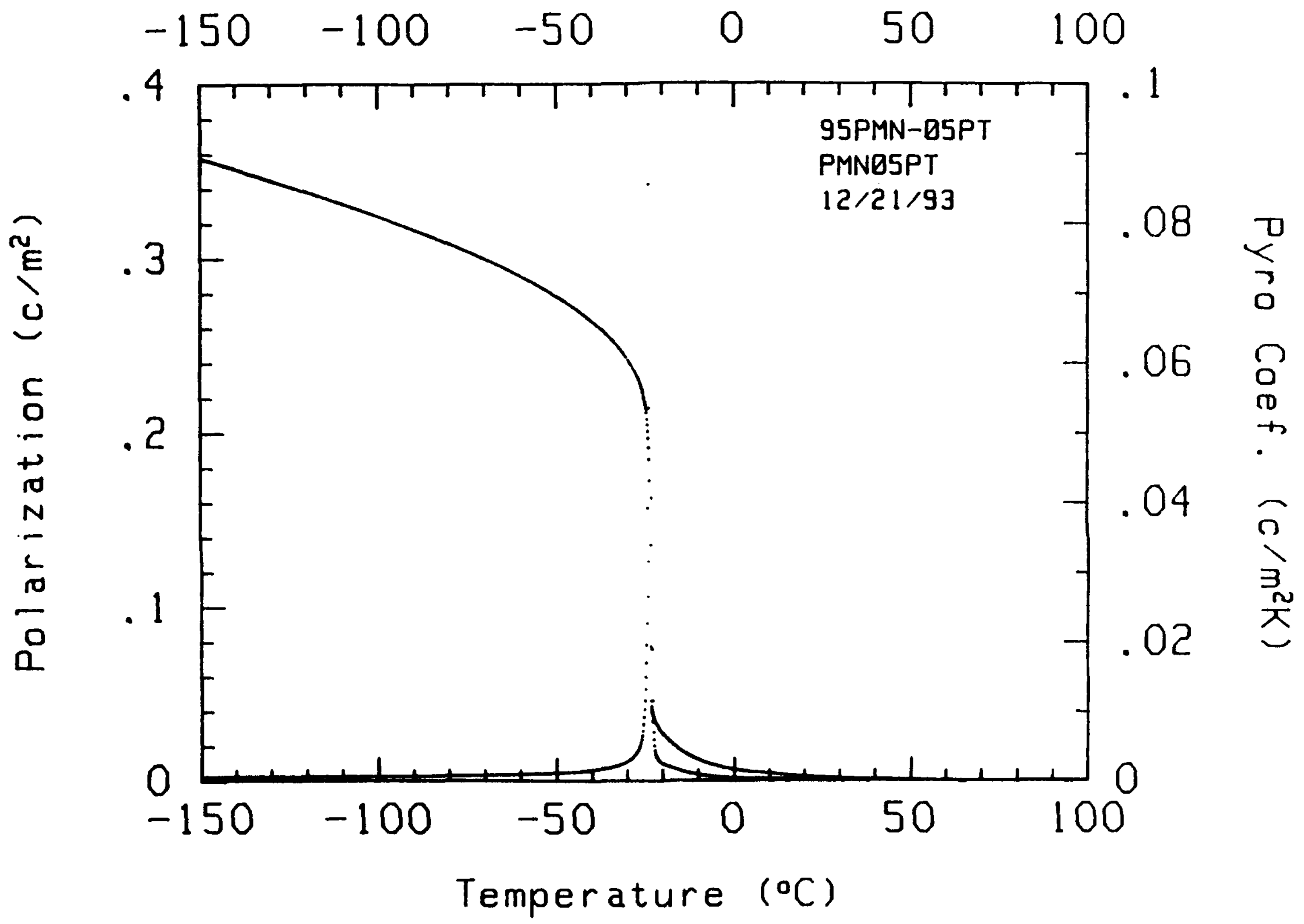
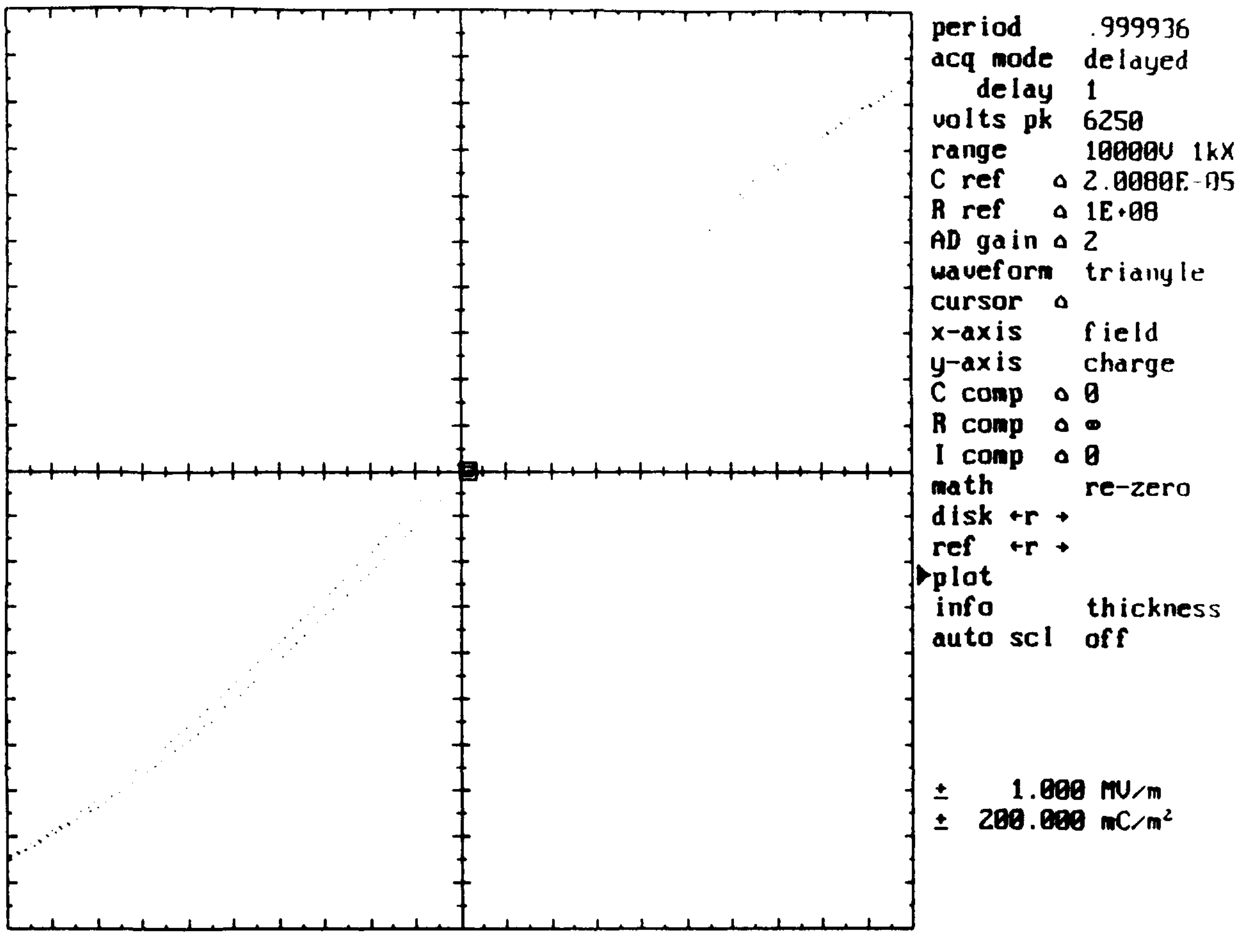
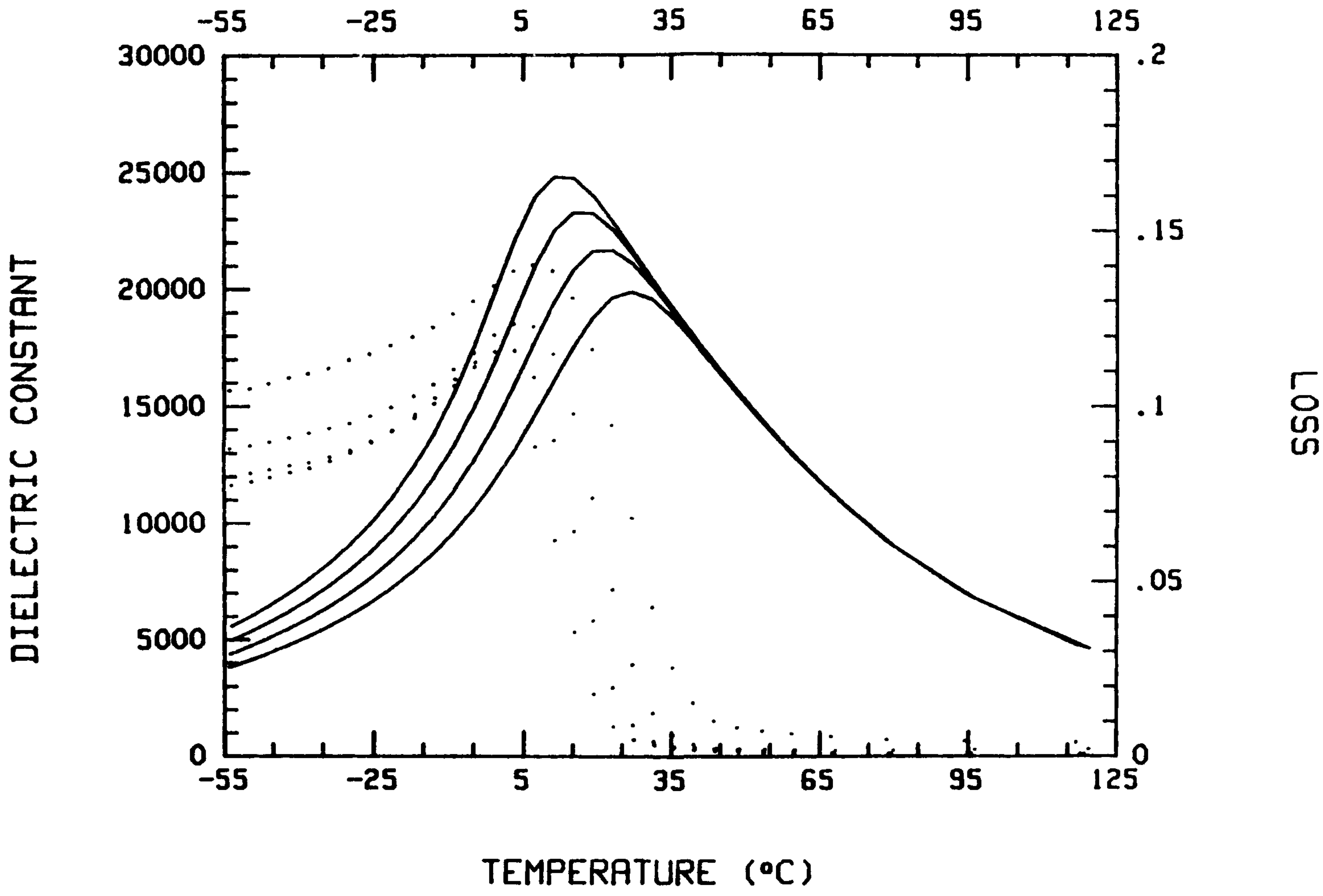


Figure 2b. Pyroelectric (dotted)/polarization (solid) temperature behavior for 95/5 PMN:PT. Note: T_d is designated at pyro peak temperature and/or steepest slope of P/temp curve.



95/5 12.27.93
dia =14.550 mm   area= 1.663E-04 m^2 thickness=6680.000 μm

Figure 2c. Polarization - E-field behavior of 95/5 PMN:PT @ room temperature.
Note: y axis - 0.2 c/m^2 full scale
x axis - 10 kV/cm full scale



.90PMN-.10PT + 1% LA
1200C/2HRS.
12/22/93

Figure 3a. Dielectric temperature behavior for 1/90/10 PLMNT. Note: Frequencies used 100 Hz, 1 Khz, 10 kHz, and 100 kHz.

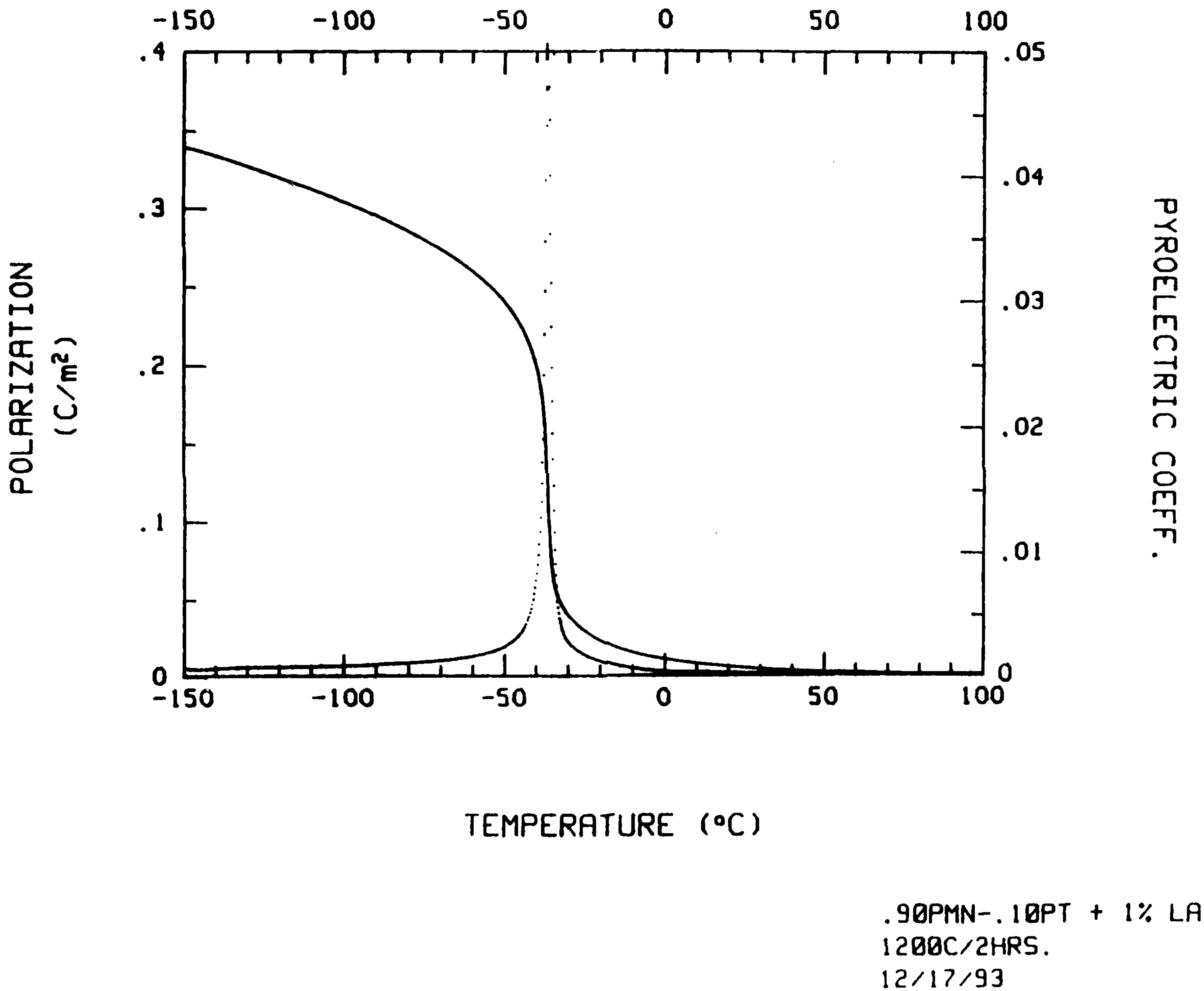
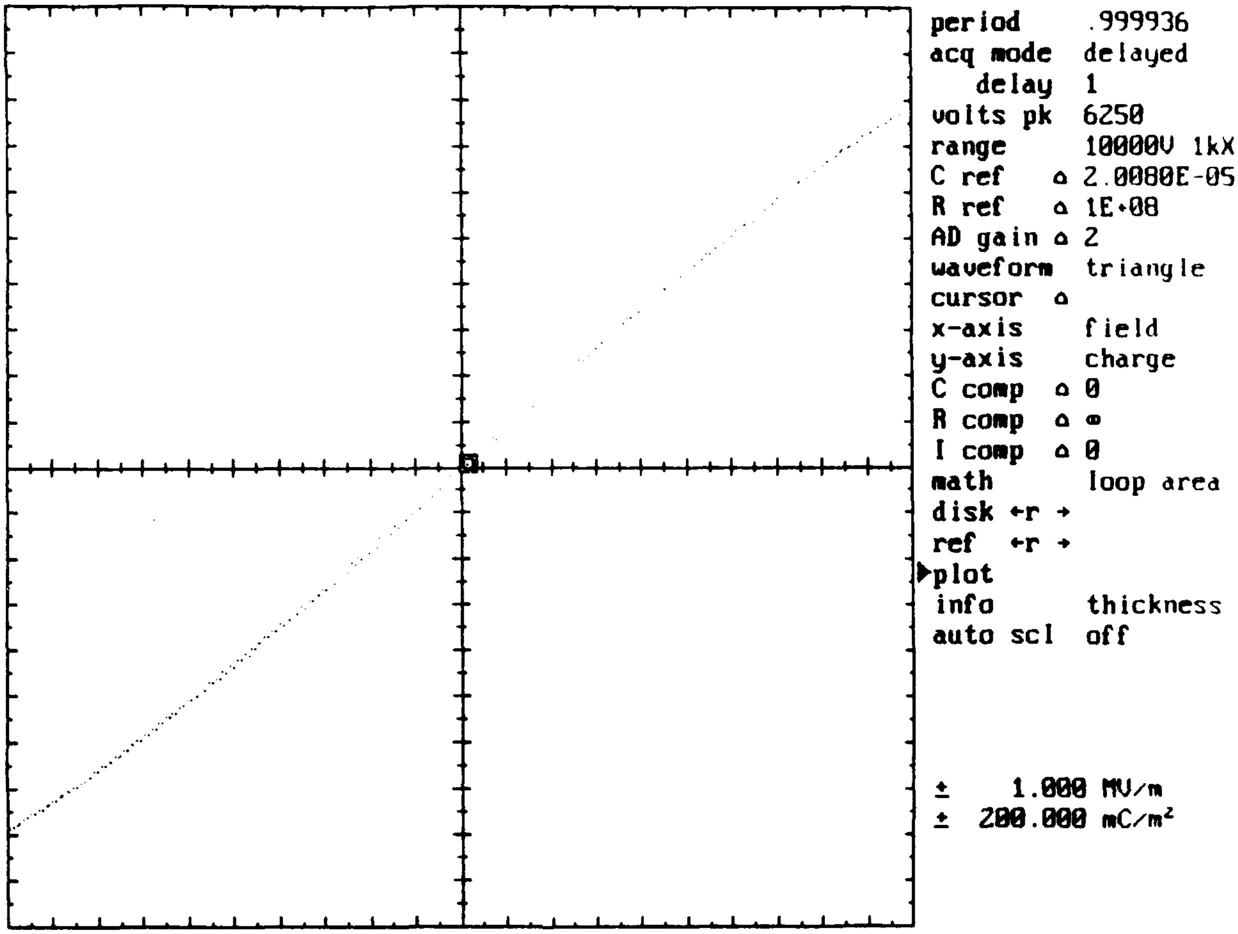


Figure 3b. Pyroelectric (dotted)/polarization (solid) temperature behavior for 1/90/10 PLMNT.



1/90/10 12.27.93
dia =14.550 mm area= 1.663E-04 m² thickness=6680.000 μm

Figure 3c. Polarization - E-field behavior of 1/90/10 PLMNT @ room temperature.
Note: y axis - 0.2 c/m² full scale
x axis - 10 kV/cm full scale

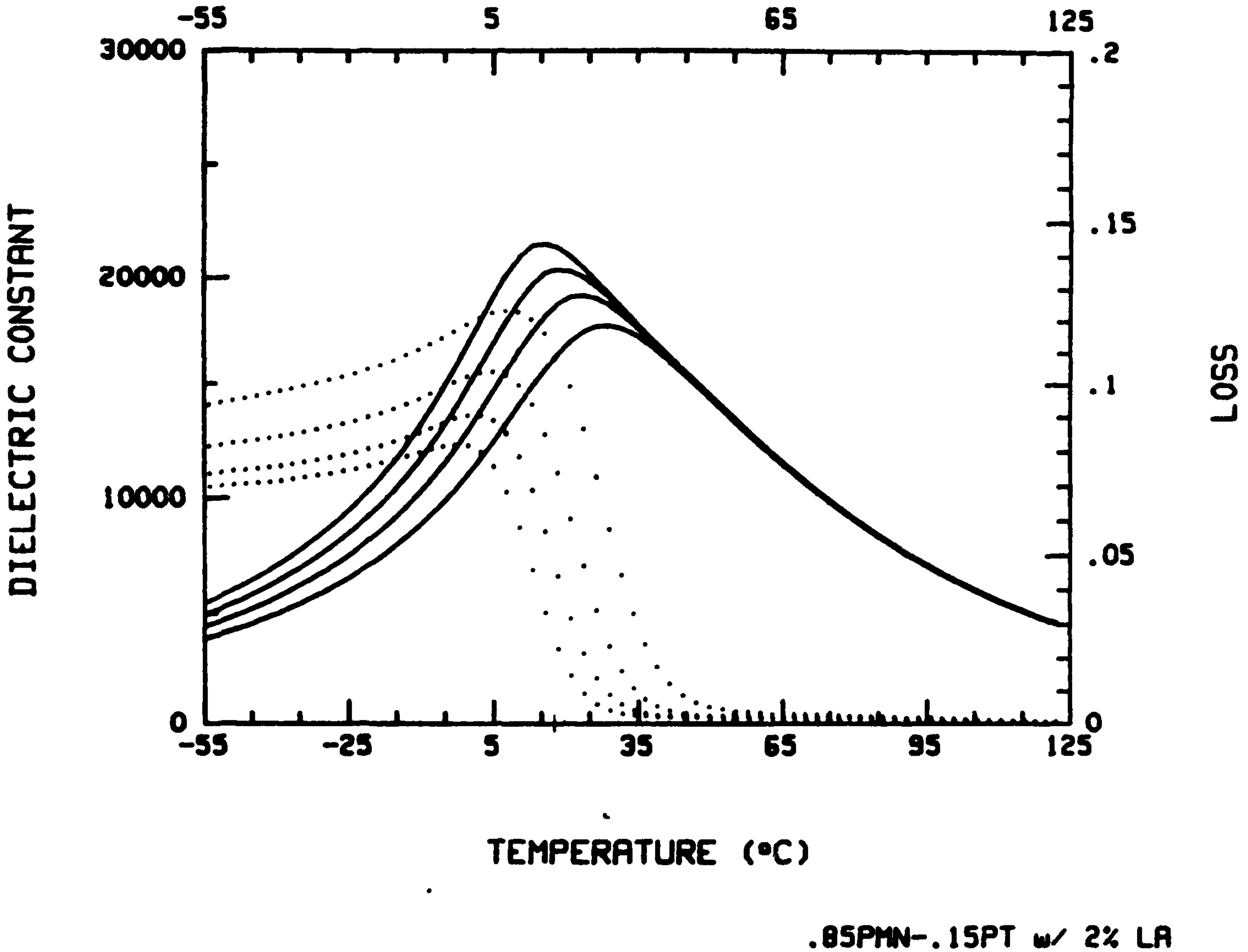


Figure 4a. Dielectric temperature behavior for 2/85/15 PLMNT.
Note: Frequencies used 100 Hz, 1 kHz, 10 kHz, and 100 kHz.

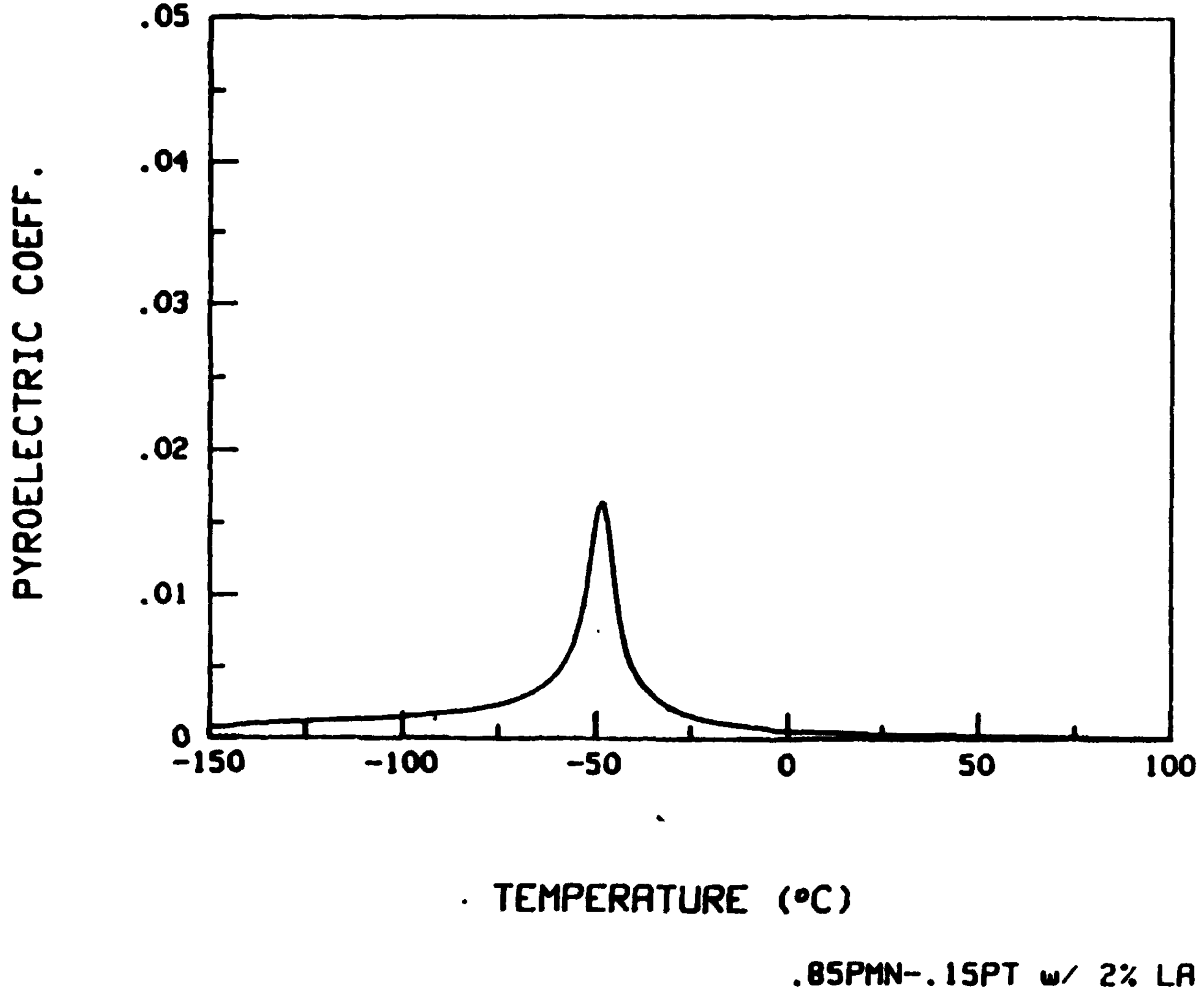
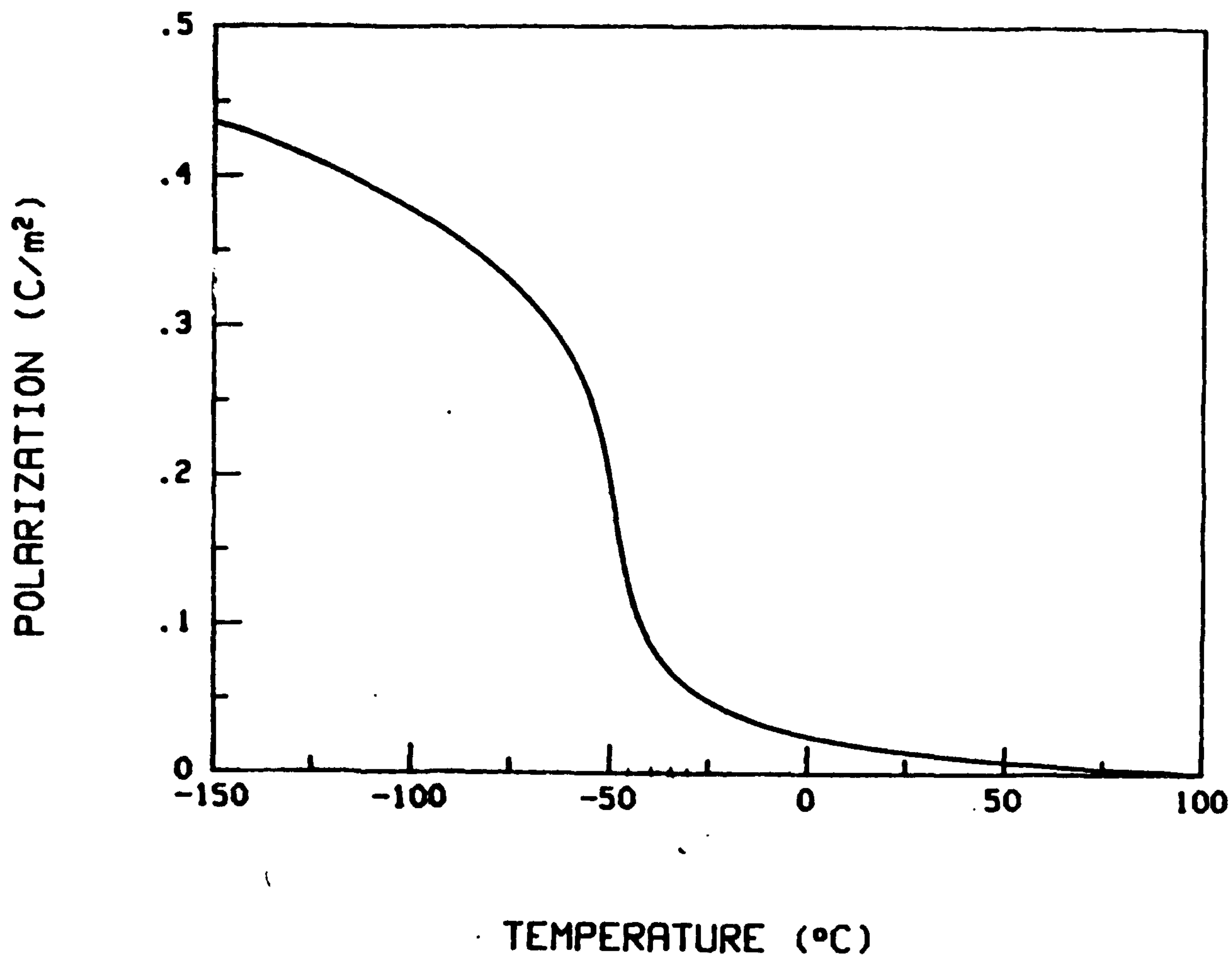
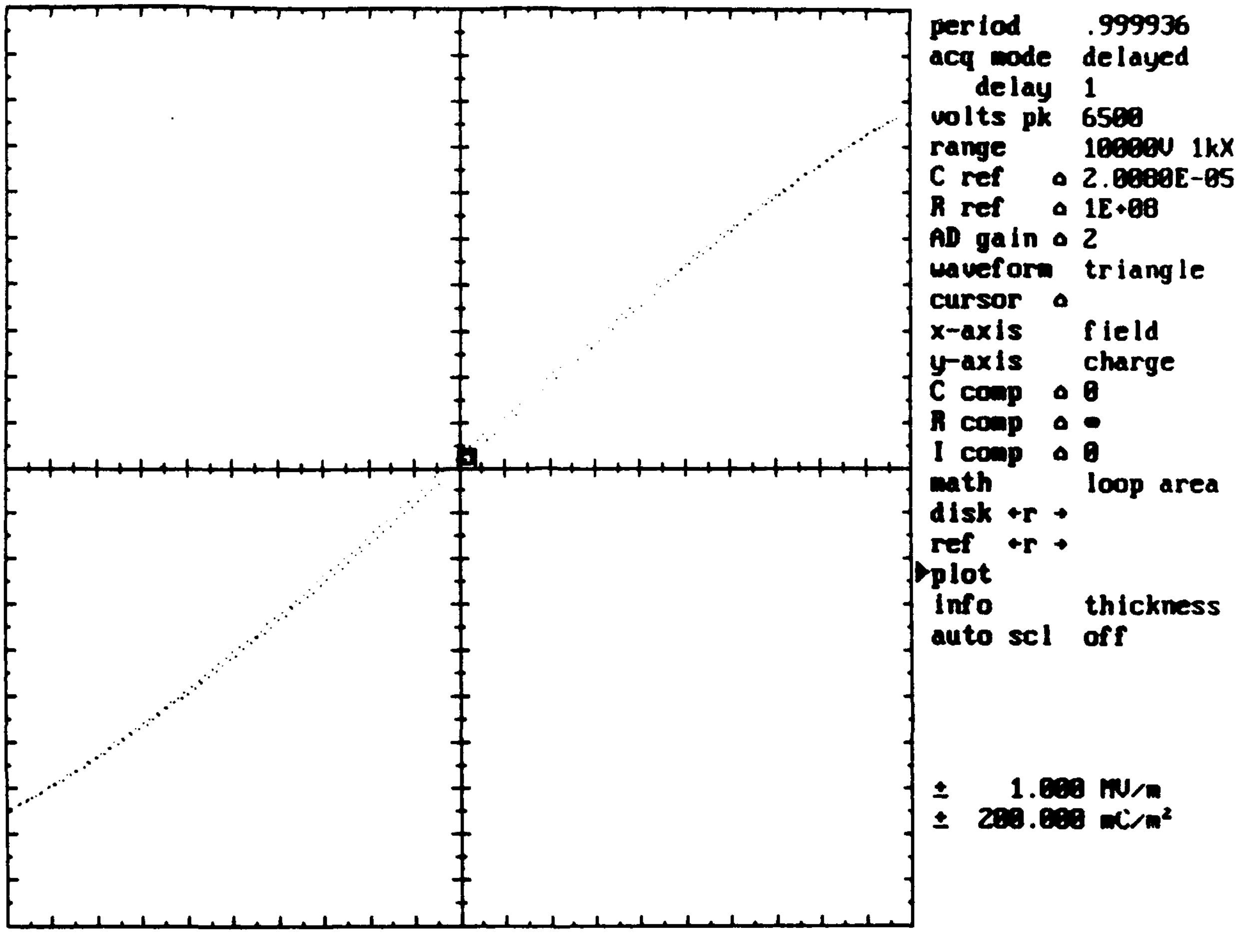


Figure 4b. Pyroelectric temperature data for 2/85/15 PLMNT. Note: Td is designated by pyro peak temperature.



.85PMN−.15PT w/ 2% LA

Figure 4c. **Polarization temperature data integrated from pyroelectric data (Figure 4b) for 2/85/15.**



85\15\2 RT SILVER
dia =14.550 mm area= 1.663E-04 m² thickness=7087.000 μm

loop 4.354

Figure 4d. Polarization E-field behavior of 2/85/5 PLMNT.
Note: y axis - 0.2 c/m^2 full scale
x axis - 10 kV/cm full scale